Time-resolved x-ray measurements of polaron dynamics of charge-ordered $Nd_{1/2}Sr_{1/2}MnO_3$

I. Kang[1], S. Johnson[1], Th. Missalla[2], A. Lindenberg[1], P. Heimann[2], Z. Qiu[1,2], J. Orenstein[1,2], H. Padmore[2], K. Kim[3], S.-W. Cheong[3], R. Falcone[1,2]
[1]University of California, Berkeley, Berkeley, CA 94720, USA
[2]Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[3]Rutgers University, Piscataway, NJ 08854, USA

The hole-doped perovskite manganese oxide materials have recently received much attention due to many fascinating phenomena such as colossal magnetoresistance (CMR). Magnetic, metal-insulator, and structural phase transitions are usually concomitant in the CMR materials owing to the strong correlations among the charge, lattice and spin degrees of freedom. We seek to attain a better understanding of the strong correlation physics of the CMR materials by time-resolving the dynamic response of electron, lattice, and spin. In this presentation, we especially address the role of the dynamic Jahn-Teller (JT) structural distortion in the phase transitions of the CMR materials.

We present the first time-resolved study of electronic and structural properties of charge-ordered $Nd_{1/2}Sr_{1/2}MnO_3$. In this system, the strong charge-lattice-spin coupling is manifested as the charge-ordered, antiferromagnetic insulating phase at low temperature ($<T_{co}$~160K). The material goes through insulator-metal, structural, *and* magnetic (antiferro-ferro) phase transitions at T_{co}. It is believed that metallicity strongly depends on the structural distortion around Mn^{3+} ions due to the JT polarons. Below T_{co}, the JT polarons possess a long-range spatial coherence due to the charge-ordering. Thus, the lattice structure of the charge-ordered state can be viewed as a polaron "solid" with a long-range order.

We monitored how metallicity and lattice structure evolves after the material was irradiated with 150-fs, 800-nm laser pulses. The pump-laser wavelength coincided with a known Mn^{3+} to Mn^{4+} charge-transfer transition. The electronic and lattice dynamics were probed with the time-resolved optical reflectivity and x-ray Bragg diffraction, respectively. The temporal resolutions were limited by the duration of the probe sources: 150 fs for optical reflectivity at 800 nm, ~80 ps for time-resolved x-ray diffraction at Beamline 7.3.3 of the ALS.

In these measurements, we observed ultrafast photo-induced metallization and structural modification of the charge-ordered $Nd_{1/2}Sr_{1/2}MnO_3$. In the transient optical reflectivity measurements, we observed an instantaneous decrease of the reflectivity, followed by slow recovery (~100 ps). The magnitude of the decrease was strongly correlated with the phase of the material: larger than 20% at the insulating (charge-ordered) phase, while ~1% at the metallic phase. Considering the fact that the equilibrium reflectivity of the metallic phase is ~40% less than that of the insulating phase, the observation is consistent with the transient charge-delocalization of the charge-ordered states due to the pump-induced charge transfer. In the time-resolved x-ray diffraction, the diffracted intensity of the (121) peak of the charge-ordered state decreased by ~20% within 80 ps after the pump laser pulses, which is limited by the ALS pulse duration. The diffracted intensity recovered in 100-200 ps. The observed lattice dynamics can be explained by the melting of the lattice of polarons. The charge transfer by 800-nm laser light produces itinerant electrons, which destroys the long-range order of the polarons. However, the electrons will eventually be trapped in Mn^{+3} sites, again forming polarons. Thus, the process can be viewed as ionization of polarons into Mn^{+4} and itinerant e_g electrons and recombination of those into polarons.

In conclusion, we report the time-resolved x-ray measurements of polaron dynamics of charge-ordered $Nd_{1/2}Sr_{1/2}MnO_3$, which clearly illustrates the strong coupling between the charge and lattice in this system.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory; additional support was provided by Laboratory Directed Research and Development funds from Lawrence Berkeley National Laboratory.

Principal investigator: Roger Falcone, Physics, University of California, Berkeley.
Email: rwf@physics.berkeley.gov. Telephone: 510-642-3316.